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HIGHLIGHT

Outsmarting Waveguide Losses in Thin-Film Light-Emitting Diodes

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Thin-film organic light-emitting diodes (OLEDs) suffer from poor external efficiencies due to—among other things—waveguide losses.[1] In the past, there have been several attempts to overcome this problem and to spatially and spectrally redistribute the electroluminescence. A new approach is now presented by Tsutsui and his colleagues.[2] They introduce an aerogel interlayer to double the external efficiency of their devices. This article will give a short overview about activities in this direction (Fig. 1, Table 1). We will restrict ourselves to outline the concepts without giving quantitative details, since the improvements depend strongly on the detailed device structure. Instead, the reader is referred to the original literature.

Inorganic LEDs have been around for many years and have recently reached very impressive performance levels. However, they are not useful for portable display applications with the ever increasing demand for higher information content. So far, this market has been dominated by liquid crystal displays (LCDs). A new display technology is currently pushing forward, namely LEDs based on π-conjugated organic materials, offering the advantages of higher brightness, visibility from all viewing angles, and faster switching speed compared to LCDs.[3]

In OLEDs, light is generated by radiative recombination of so-called "excitons", i.e., emitters in their excited state. Unlike in conventional fluorescence spectroscopy (*optical* excitation), these excitons are generated *electrically* by combining "electrons" and "holes", which have been injected previously from the contacts. The emission color is determined by the optical "gap" between the highest occupied and lowest unoccupied orbital (HOMO and LUMO, respectively) of the emitter. In order to allow the generated light to emerge from the front surface of the device, one of the contacts is transparent (typically indium tin oxide, ITO). The total thickness of the organic layer(s) is generally around 100 nm, i.e., too thin to mechanically support itself. Therefore, an appropriate transparent substrate (glass or inert polymer, several hundreds of micrometers thick) is required. A cross-section of a typical OLED structure is depicted in Figure 1a.

Ultimately, the photons have to travel from the emissive layer ($n_{organics} \approx 1.6$–1.7) through the ITO contact ($n_{ITO} \approx 1.6$–2.0) and the substrate ($n_{substrate} \approx 1.5$) and finally into air ($n_{air} = 1$). Thus, because of total internal reflection, which occurs when going from high- to low-index media, light is trapped inside the device.

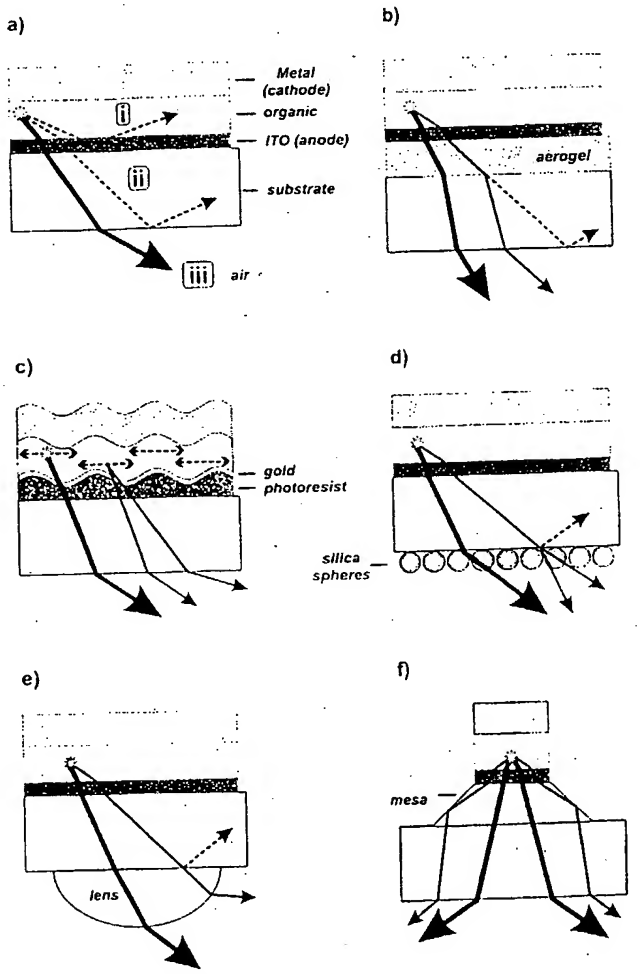

Fig. 1. Illustration of the various OLED device structures: a) "standard" structure, b) with aerogel interlayer [2], c) with integrated DFB grating [5], d) with scattering microspheres [8], e) with microlenses [9], and f) built atop a mesa structure [10]. Rays ultimately leaving the device in forward direction are printed as thick solid lines, those internally reflected as dotted lines, and those which are extracted by the respective method as thin solid lines.

Depending on the angle of light propagation, light is either i) waveguided and/or absorbed in the organic/ITO layer(s), ii) waveguided and/or absorbed in the substrate *and* the organic/ITO layer(s), or iii) coupled out in the forward direction (Fig. 1a). Simple ray optics allows the estimation of how much light will be observable in forward direction:[1]

$$\eta_{forward} \approx 0.5/n^2 \quad (1)$$

Here, n is the highest refractive index within the structure, typically the organic layer(s) or the ITO. It is assumed that the

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emission is isotropic, i.e., that there are no absorption or scattering losses, that all layers are optically thick, and that the light emitted in the backward direction is ideally reflected forward by the metal contact. Finally microcavity effects (see below) are neglected. According to Equation 1, for $n_{organics} \approx 1.6$–1.7 the total amount of light coupled out of the front surface would be only about 20 % of the generated photons. To recover at least part of the remaining 80 % has been the focus of intense research efforts over the past years. The activities can be roughly divided into the following two categories.

The first category deals with interface modifications: by introducing scattering features light that is otherwise guided inside the device can be partly extracted. We distinguish random scatterers, obtained for example by surface roughening,[4] and those with periodic structure such as distributed feedback (DFB) gratings (Fig. 1c)[5] and two-dimensional photonic bandgap structures.[6] DFB gratings have so far been realized in one dimension but the next step to two dimensions, already achieved in the context of optically pumped lasing,[7] seems logical. The introduction of scattering silica spheres (Fig. 1d)[8] is close to a periodic structure but with some degree of disorder. In all cases, the pitch of the structure determines the forward scattered wavelength, and there is a distinct dependence of the color on the scattering angle for the case of silica beads even on the position of the observer.[8] The location of the scattering layer, i.e., whether it is in contact with the organic layer(s) or the substrate, determines which waveguide modes are extracted. In some cases polarization issues also need to be considered.[5]

Attaching micro-optical elements such as microlenses to the front surface (Fig. 1e)[9] also led to improved forward output by extracting light that is guided in the substrate. Alternatively, Gu et al. built the OLED structure atop of "mesas" (truncated cones, Fig. 1f),[10a] which reflect photons initially emitted at large angles towards the surface. Further improvement of the mesa-based OLEDs was achieved by depositing a thin layer of the high-refractive-index TiO_2, which completely eliminated waveguiding in the organic and ITO layers.[10b]

The second category to improve the forward light output takes advantage of the microcavity structure of OLEDs: since the emissive material is sandwiched between two conductive layers with increased reflectivity, this structure acts like a microcavity, a photonic structure with at least one dimension on the order of optical wavelengths.[11] Even in the case of "normal" OLEDs, using a poorly reflecting ITO electrode, the spontaneous emission characteristics are altered. However, in many cases these microcavity effects are "hidden" because of the broad bandwidth of the emission spectra typically observed for most organic compounds ($\Delta\lambda$ = 50–100 nm).[11b]

Replacing the ITO by a semitransparent metal layer or depositing ITO onto a dielectric mirror allows the characteristics of the microcavity to be adjusted within a wide range. The cavity changes the spectral linewidth (narrower than in free space), the direction (directed along the optical axis of the cavity, i.e., in forward direction), and the lifetime.[11] Therefore, microcavities have been used to investigate lasing effects in organic materials.[12] Another important parameter is the thickness of the organic layer(s).

Tsutsui et al. report on a new way of improving the external efficiency in OLEDs in the latest issue (No. 15) of *Advanced Materials*.[2] They take advantage of the fact that the organic layer(s) and the ITO contact are rather thin, i.e., they represent rather poor waveguides with only very few allowed modes. Therefore, a considerable amount of light leaks into the substrate, and Equation 1 no longer holds. If additionally the index of refraction of the substrate is low, the light output can be improved.[2] The only problem so far was the lack of a suitable substrate material. Tsutsui et al. now propose the use of aerogels with a refractive index close to that of air.[2] It is an important achievement to make this idea compatible with typical OLED fabrication techniques by introducing the aerogels as an interlayer between an ordinary glass substrate and the OLED itself (Fig. 1b). This way, the glass substrate acts just like an ordinary glass window, introducing a simple offset in the light path. Therefore, emission from the substrate's edges is not observed unlike in standard OLEDs (Fig. 1a). Also, it is important to use a hydrophobic aerogel, because the hydrophilic kind would lead to fast degradation. Using this new and exciting approach the external efficiency was doubled.

So far, we have discussed "physical" modifications of the device structure; however, this article would not be complete without pointing out "chemical" contributions, for example, by taking advantage of anisotropic emission, such as in a liquid crystalline environment. Kim et al.[13] predicted from their theoretical half-space model an approximately twofold increase of the external efficiency for in-plane emission over isotropic emission. One approach would be to use a nematic/smectic LC-phase with in-plane alignment, an opportunity widely recognized to achieve linearly polarized emission.[14] Researchers working in this direction have focused on improving the polarization ratio; however, little attention has been paid to the effect of improved light output. On the other hand, the use of a discotic LC-phase with perpendicular alignment of the columns would be ideal, since the emitting disks could then be oriented within the plane of the substrate and all light would be emitted in the forward direction. There are only very few reports on OLEDs based on discotic emitters.[15] The authors point out a reduction of onset fields when going from the isotropic phase to the mesophase,[15a] but unfortunately possible changes of the device efficiency are not considered. This approach bears the disadvantage that strong intermolecular coupling often observed in π-stacks may lead to a strong reduction of the luminescence efficiency.[16]

In conclusion, a number of "physical" concepts have been successfully applied to outsmart waveguide losses in OLEDs, while "chemical" means have so far only played a minor role. Improvements of the overall light output by factors of 2–5 (some even >10 for normal emission) have been reported. However, not all of the proposed methods are useful for display applications, some may be more applicable to large-surface light sources (all concepts based on scattering). The new method by Tsutsui et al.[2] is very promising because of its simplicity; however, the technical feasibility of the aerogel process as well as the long-term robustness of the devices still have to be tested. It is further noteworthy that by incorporating the

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aerogel the emission characteristics remain unchanged. The same applies to surface roughening but this technique usually produces a much smaller improvement over the standard structure. All other methods somehow influence the emission characteristics, e.g., polarization, direction, color, etc. (see Table 1), which may or may not be desirable in some applications.

Table 1. Overview of the concepts to improve the forward light output from thin-film OLEDs.

Method	Effects on [a]			Extracting light from [a]			Ref.
	Polarization	Directionality	Spectrum/ color	Substrate	Organics/ ITO	Thickness dependence [c]	
Substrate Refractive Index		- [c]	-	y	-		[2]
Interface Modifications							
Roughening		-	-	y		-	[4]
Diffraction	y	y	y	-	y	y	[5]
Photonic bandgap [b]	y	y	y	y		y	[6]
Microcavity [c]		y	y	y	-	-	[8]
Microlens		y		y	-	-	[9]
Metal		y		y	-	-	[10]
Metal grids		y		y	y	y	[11]

[a] Comparison to the standard structure (Fig. 1a) without any of the listed features. [b] Diffraction at a periodic scattering feature is deposited onto the substrate on the transparent electrode contact with the organic layer. [c] Thickness of the organic layer.

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